73

LA-UR- 01 01-4289

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Title: IMPROVED EVALUATION OF 239PU (N,F) BETWEEN 0.1 AND 20 MEV INMCIDENT NEUTRONS ENERGIES

Author(s): P. Talou, T-16, LANL
P.G. Young, T-16, LANL
M.B. Chadwick, T-16, LANL

Submitted to: American Nuclear Society - Accelerator Applications Division Conference
"Nuclear Applications in the New Millenium," Reno, NV - Nov. 11-15, 2001

Improved Evaluation of ^{239}Pu (n,f) between 0.1 and 20 MeV Incident Neutrons Energies

P. TALOU*, P.G. YOUNG, AND M.B. CHADWICK

T-16, Nuclear Physics Group,
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

Abstract-Accurate cross sections lie at the heart of successfull neutronics simulations. The advent of innovative nuclear designs such as Accelerator Driven Systems (ADS) have recently emphasized the need for accurate measurements, evaluations, and calculations of neutron-induced fission cross sections over a wide range of nuclei and energies. As a first step, we have performed a thorough covariance analysis of the neutron induced fission cross section of ^{239}Pu between 0.1 and 20 MeV. The choice of this first study has been driven by the importance of this Pu isotope in the US nuclear waste stream. Newly available experimental data (both absolute and in ratio to the standard ^{235}U) have been included in this new evaluation. A Bayesian statistical approach has been used to infer posterior knowledge on the cross sections and on the associated errors (standard deviations + correlations). Significant reductions of these errors are observed, compared to the previous ENDF/B-VI evaluation. Large changes (up to 4% in places) appear above 14 MeV incident neutron energies, mainly due to a recent revised ^{235}U (n,f) evaluation. Overall very good agreement is observed elsewhere. Finally, a comparison between this new evaluation and other existing evaluations is discussed.

I. INTRODUCTION

Driven by the amazing progress of modern particle accelerators capabilities, several new nuclear applications are emerging. Among them, the transmutation of nuclear waste [1] and the safer accelerator-driven civil nuclear reactor designs [2] are tremendously important. While the planet faces more and more environmental threats due to previous and current energy policies in the World, the energy extracted from the atomic nucleus could very well play a key role in the near future energy resources. Of course, old worries relating to the use of nuclear power have not disappeared: the danger of a major nuclear reactor accident, the long-time uncertainties of waste disposal, the associated worries of military "Plutonium mines", are all potential threats which need to be addressed before any new nuclear energy policy has any chance to be widely accepted. This is exactly where such new Accelerator-Driven Systems (ADS) come into play by proposing improved and safer solutions to most of these problems.

Such new nuclear designs call for accurate nuclear data libraries not necessarily available today [3]. Most existing data libraries have been developed for existing applications and therefore do not necessarily meet the expectations of upcoming projects. In this context, the nuclear physics group in Los Alamos has started a project to extend existing nuclear data libraries to fulfill requirements for the ATW (Accelerator Transmutation of Waste) program in an international collaboration context.

Part of this work includes covariance analyses of various isotopes important for ATW. They can be important as constituent of the actual waste fuel or as part of the shielding or/and target design. Accurate cross sections are then crucial to gauge safety issues as well as efficiency (burn-up) factors. Large discrepancies among current data libraries exist for instance in minor actinides like Cm, Am and Np isotopes. These discrepancies strongly influence the effective neutron multiplication factor k_{eff} as well as its evolution in time, as revealed by a recent ATW simulation [4]. These large uncertainties need to be resolved before any ATW burner prototype can be built, safely and efficiently.

This paper focuses on the evaluation of the cross section of neutron induced fission of Plutonium-239, ^{239}Pu (n,f), for incident neutron energies between 0.1 and 20 MeV. Because of the large quantity of ^{239}Pu present in the US nuclear waste stream, this isotope is of primary importance for our purpose here. Also, new experimental data sets have recently become available, and therefore encouraged us to reevaluate this reaction, as a starting point in our more general evaluation program.

*talou@lanl.gov

This paper is organized as follows. The next section briefly reviews the mathematical framework used in our data evaluations, namely the Bayesian statistical inference scheme. The following section gathers our new results and a discussion of its validity, along with a comparison with the new japanese evaluation JENDL-3.3. Finally, the conclusion summarizes the main results obtained in the present work.

II. BAYESIAN INFERENCE SCHEME: A BRIEF REVIEW

Most modern data evaluations make use of a famous theorem developed by Th.Bayes in 1764 based on conditional probabilities theory. Bayes' theorem provides a rule for updating the belief in a given hypothesis H (i.e., the probability of H being true), given new acquired knowledge D, and some circumstances C:

$$P(H|D,C) = \frac{P(D|H,C)P(H|C)}{P(D|C)}, \quad (1)$$

The left-hand term $P(H|D,C)$ is called the ***posterior***, and represents the new belief in the hypothesis H after gaining the new knowledge D. The term $P(H|C)$ is the ***prior*** probability of H given C alone. Finally, the term $P(D|H,C)$ is the ***likelihood*** function which gives the probability of observing D if the hypothesis H and the circumstances C were actually true. The denominator $P(D|C)$ is independent of H and can be regarded as a normalizing constant. In other words, the Bayes' rule reads simply

$$\text{Posterior} \propto \text{Likelihood} \times \text{Prior} \quad (2)$$

Such a rule can then be applied iteratively each time new information becomes available, while keeping in "mind" all the previous gathered informations. The eventual correlations between different pieces of informations are automatically considered and taken into account in the determination of the posterior. Such an iterative scheme precisely illustrates the philosophical concept of ***inference.***

Priors have been the subject of intensive specialized literature and controversy. Nevertheless, a convergence of opinions seem to have emerged recently. When data (i.e., information) are prolific, then any prior is suitable and will not influence the final evaluation of the posterior. On the other hand, when data are scarce, the maximum entropy principle should be used to infer appropriate priors. Applying Bayes' rule of inference in this context leaves no place for doubt or imprecision, at least for its mathematical framework. The correct estimates of experimental uncertainties is another matter. It is not rare to find two or more *inconsistent* data sets; even when data are not inconsistent, it is usually quite difficult to obtain reasonable estimates of experimental error bars. Most divergences between existing data libraries arise purely from this bare fact.

It is not our purpose here to develop at length the mathematical details of the Bayesian inference scheme; therefore we prefer to orient the reader toward already existing good literature on the subject- see for instance [5] and [6]. Such a Bayesian approach has been implemented in the numerical code GLUCS, "a Generalized Least-Squares Program for Updating Cross-Section Evaluations with Correlated Data Sets", developed by D.M. Hetrick and C.Y. Fu from OakRidge National Laboratory [7]. We used this particular code for the present evaluation. For details on the coding, please refer to the GLUCS user manual [7].

III. NEW ^{239}PU (N,F) EVALUATION: RESULTS AND DISCUSSION

The set of experimental data we used here differs in several respects from the one used in the last ENDF/B-VI evaluation. First, not all of the data sets used previously were included. Indeed, some relatively older experiments exhibit large uncertainties in the cross sections. While such results could be included carefully when data is scarce, the present amount of available data allows to safely neglect them. On the other hand, several new data sets have been included in the new version. These data come from either recent experiments performed later than 1990, or data which were not present in the EXFOR database by the time the last ENDF evaluation was performed; this especially concerns data from Russia. Our complete database is gathered in Tables 1 and 2, for absolute and ratio to ^{235}U (n,f) data respectively.

Our new ^{239}Pu (n,f) evaluation is plotted in Fig. 1 for incident neutrons energies between 0.1 and 20 MeV, along with the current ENDF/B-VI evaluation. Two evident conclusions can be drawn from this figure: (1) there is an overall very good agreement between the two evaluations for energies below 14 MeV; (2) large discrepancies appear above 14 MeV. The observed differences can be as high as 4% in places. We will see that such a big difference is due to a revised version of the ^{235}U (n,f) evaluation used here versus the one used earlier.

In Fig. 2 are plotted the calculated standard deviations for the two evaluations. A significant improvement on these errors emerge from our new evaluation. Cross section of ratio evaluation ^{239}Pu (n,f) / ^{235}U (n,f) is plotted on Fig. 3, along with the two most recent experimental data sets from P.Staples et al. [8] and O.Shcherbakov et al. [9].

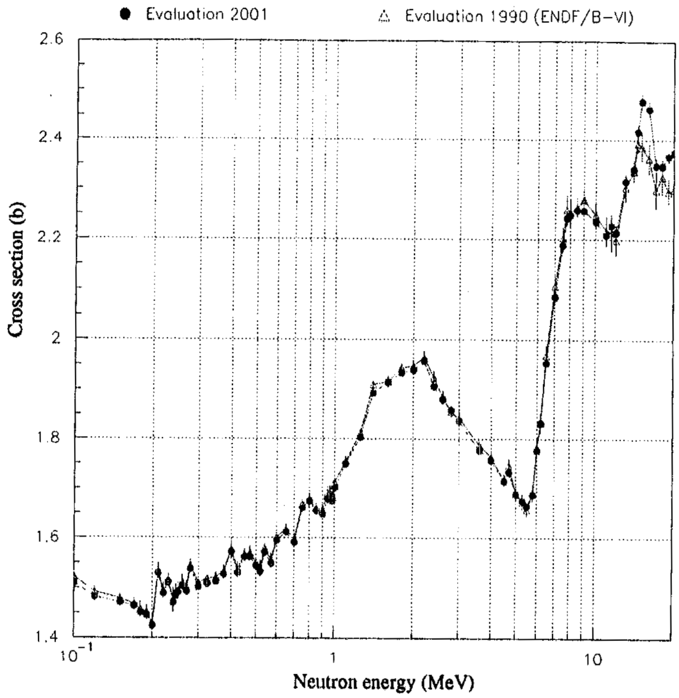

FIG. 1. New ^{239}Pu (n,f) evaluation for incident neutrons energies between 0.1 and 20 MeV. This new evaluation is compared to the current ENDF/B-VI evaluation.

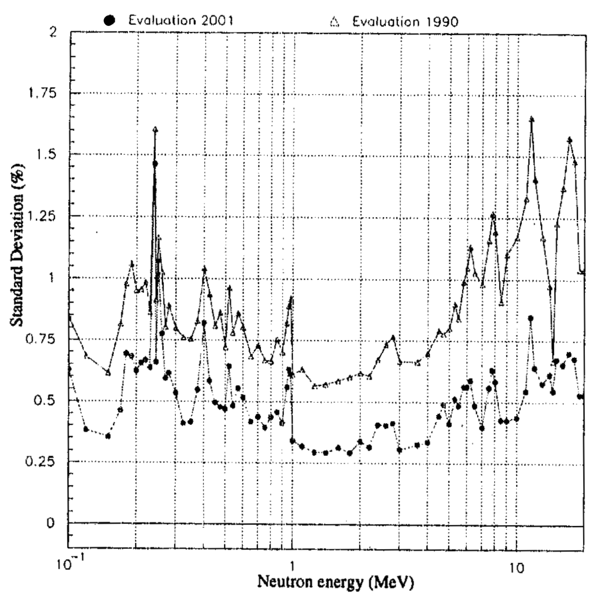

FIG. 2. Standard deviations associated with the two evaluations plotted in Fig. 1.

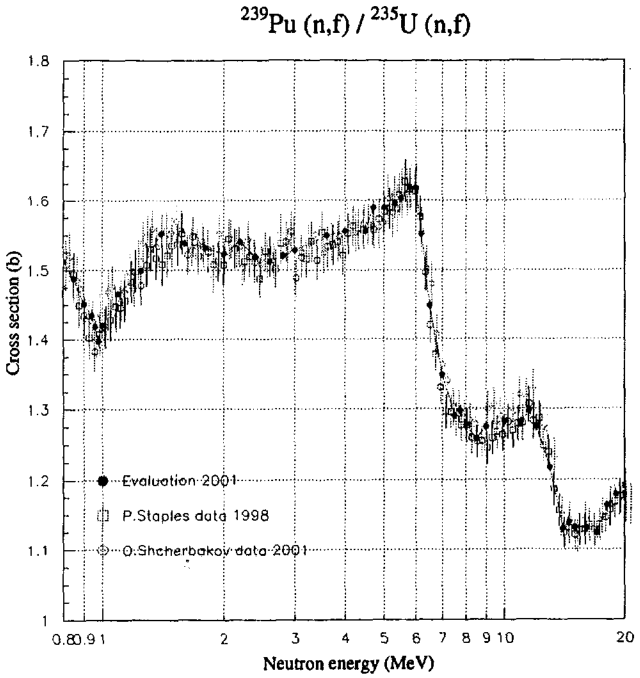

FIG. 3. Evaluation of the ratio cross section ^{239}Pu (n,f) / ^{235}U (n,f), along with the two most recent experimental data sets from P.Staples et al. [8] and O.Shcherbakov et al. [9].

Finally, it is very instructive to compare our present evaluation with the newly released Japanese evaluation JENDL-3.3 [10]. This is summarized on the Fig. 4. Large discrepancies appear in places, clearly drawing some inconsistencies between JENDL and ENDF. Specifically, the 1-2 MeV region shows a large deviation of ENDF vs. JENDL. Actually, this difference is almost entirely contained in the cross section used for ^{235}U (n,f), as can be seen on Fig. 5. As such an evaluation is considered a *standard* for both JENDL and ENDF, resolving such large discrepancies is a very high priority for future evaluations.

IV. CONCLUDING REMARKS

We have performed a new evaluation of the cross section of neutron induced fission reaction on ^{239}Pu, for incident neutron energies between 0.1 and 20 MeV. This work is part of a larger effort toward a modern evaluation of all isotopes important for the ATW project, which aims at reducing significantly the danger of nuclear wastes disposal. ^{239}Pu appears in very large quantities in the US nuclear waste stream and therefore constitutes a reasonable first choice for our evaluation program.

The Bayes' theorem of conditional probabilities lies at the heart of this new evaluation. This powerful tool allows an increase of knowledge, step-by-step, as experimental data become available. Such a technique aims at reducing systematic errors unavoidable in a given experimental setup. While the mathematical framework of such a technique appears sound, reliable estimates of experimental errors still constitutes the main challenge of any reasonable data evaluation.

The present evaluation uses most of the experimental database established for the previous ENDF/B-VI evaluation, with some differences though. Some older data with large uncertainties were not included in the current evaluation. Also, newly available experimental data sets have been included.

The comparison of the new versus the older ENDF evaluations carry a few comments. First of all, there is an overall very good agreement between the two evaluations below 14 MeV incident neutrons energies. Associated standard deviations are nevertheless significantly reduced in this new work. Large discrepancies in the (n,f) cross section appear above 14 MeV. This is mainly due to the use of a revised version of the standard ^{235}U (n,f) above this exact same energy.

A comparison of this new ENDF evaluation and the recently available Japanese evaluation JENDL shows some large discrepancies in places. That is the case for incident neutrons of 1-2 MeV, where big differences are mainly driven by the use of two significantly different standard ^{235}U (n,f) evaluations. We should note that these differences were also present in previous evaluations. As such a reaction is a standard upon which many other evaluations are established, we think that resolving this discrepancy should be of highest priority.

ACKNOLEDGMENTS

The authors would like to thank O. Shcherbakov for providing his experimental data points prior to publication, and T.Kawano for valuable discussions concerning discrepancies between JENDL-3.3 and the present evaluation.

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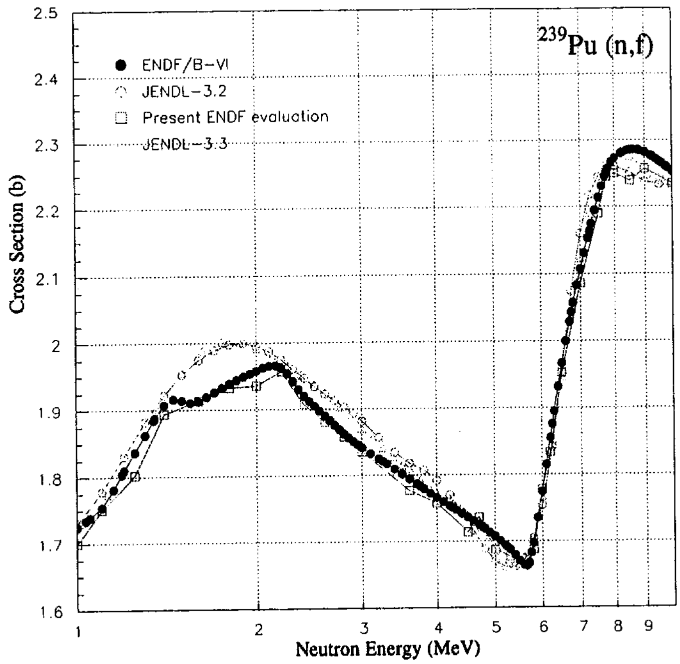

FIG. 4. Comparison of several existing evaluations of ^{239}Pu (n,f).

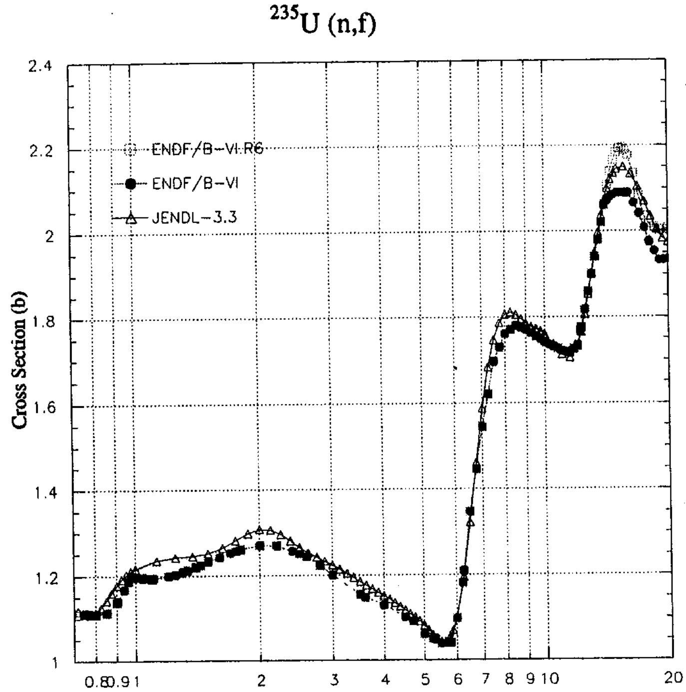

FIG. 5. ^{235}U (n,f) evaluations from different libraries. The ENDF/B-VI.R6 evaluation (squares) is a recent revision of the ENDF/B-VI evaluation above 14 MeV, taking into account data from P. Lisowski [11].

APPENDIX A: EXPERIMENTAL DATABASE

CSISRS #	First Author	Institute	Year	Energies
10267-2	R. GWIN	USA-ORL	1976	5-200 keV
10545-2	M.C. DAVIS	USA-MHG	1978	0.1-1 MeV
12826-3	M. MAHDAVI	USA-MHG	1982	14.63 MeV
20476-3	M.G. SCHOMBERG	UK-HAR	1970	0.1-30 keV
20567-3	I. SZABO	FR-CAD	1976	35 keV - 1 MeV
21704-2	C. WAGEMANS	BLG-MOL	1980	thermal - 30 keV
22304-5,9	K. MERLA	GER-DRE	1991	~MeV
30475-5	R. ARLT	DDR-TUD	1981	14.7 MeV
30634-3	LI JING-WEN	CPR-AEP	1982	14.7 MeV
30670-2	ZHOU XIAN-JIAN	CPR-AEP	1982	1-5.6 MeV
30706-4,5,6	C.M. HERBACH	DDR-TUD	1985	4-20 MeV
40470-3	A.A. BERGMAN	CCP-JIA	1976	0.2-30 keV
40547-8,9	V.M. ADAMOV	CCP-RI	1977	1.42 & 14.8 MeV
40911-7	I.D. ALKHAZOV	CCP-RI	1983	14.7 MeV
40927-5	I.D. ALKHAZOV	CCP-RI	1986	1-20 MeV

TABLE I. ^{239}Pu (n,f) absolute cross section data. The column labeled CSISRS represents the data entry number in the Brookhaven National Nuclear Data Center CSISRS experimental data compilation.

CSISRS #	First Author	Institute	Year	Energies
10086-4	W.P. POENITZ	USA-ANL	1970	0.15-1.4 MeV
10253-2	W.P. POENITZ	USA-ANL	1972	0.03-5.29 MeV
10562-2	G.W. CARLSON	USA-LRL	1978	1-30 keV
10734-2	J.W. MEADOWS	USA-ANL	1978	0.1-10 MeV
12766-2	L.W. WESTON	USA-ORL	1984	~kev - 0.2 MeV
13134-9	J.W. MEADOWS	USA-ANL	1988	14.74 MeV
13801-2	P. STAPLES	USA-LAS	1998	0.85-62 MeV
20363-3	E. PFLETSCHINGER	GER-KFK	1970	5.2 keV - 1 MeV
20409-3	S. CIERJACKS	GER-KFK	1976	0.4-20.9 MeV
20428-4	D.B. GAYTHER	UK-HAR	1975	1 keV - 1 MeV
20568-3	M. SOLEILHAC	FR-BRC	1970	0.3-1.4 MeV
20569-4	I. SZABO	FR-CAD	1976	10-200 keV
20779-6	M. CANCÉ	FR-BRC	1978	13.9 & 14.6 MeV
20786-5	K. KARI	GER-KFK	1978	1-21 MeV
30588-5	M. VARNAGY	HUN-KOS	1982	~14 MeV
40020-2	M.V. SAVIN	CCP-KUR	1969	0.82-5.35 MeV
40027-3,5	G.N. SMIRENKIN	CCP-FEI	1967	~MeV
40309-2	V.G. NESTEROV	CCP-CCP	1968	~MeV
40412-8	V.N. KONONOV	CCP-FEI	1975	10-80 keV
40476-4	K.D. ZHURAVLEV	RUS-NIR	1976	2,24,55,144 keV
40563-2	P.E. VOROTNIKOV	CCP-KUR	1979	30-200 keV
40601-4	A.A. BERGMAN	CCP-JIA	1980	0.1-40 keV
40751-7	A.A. BERGMAN	CCP-JIA	1980	0.1-50 keV
40824-2,3	B.I. FURSOV	CCP-FEI	1977	0.024-7.4 MeV
-	O. SHCHERBAKOV	Petersburg Nucl. Phys. Inst.	2001	0.6-20 MeV

TABLE II. ^{239}Pu (n,f) / ^{235}U (n,f) ratio cross section data.